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(57) Abstract

The invention concerns a lignin-based adhesive binder composition for particle boards and similar wood-based products, comprising a foamed aqueous mixture of fibers and carbohydrates obtained from a process of pulping lignocellulosic materials, the mixture being foamed to 1.1 to 10 times the volume of the liquid mixture. The invention also concerns a method of preparing a lignin-based adhesive binder, comprising the steps of providing lignin isolated from a pulping process of lignocellulosic materials, providing fibers based on wood or annual or perennial plants, forming an aqueous suspension of the lignin and the fibers, and foaming the aqueous suspension to provide a foamed adhesive binder composition containing the solids in a homogeneous dispersion.

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LIGNIN-BASED ADHESIVES FOR PARTICLE BOARD MANUFACTURE

The present invention relates to the manufacture of particle boards and similar wood-based products comprising lignocellulosic particles, fibers or flakes mixed with and bonded together with an adhesive binder. In particular the present invention concerns novel lignin and/or carbohydrate -based adhesives and a preparation process thereof as well as particle boards manufactured using the adhesives.

The rapid increase in the production of particle boards and fiber boards, especially medium density fiber boards (in the following also abbreviated MDF boards), during the last decades has created a demand for adhesives that are inexpensive, available in large quantities, and independent of crude oil. Lignin meets well these requirements, and it does not contain any formaldehyde, which traditionally been considered a serious problem with conventional urea-formaldehyde (UF) adhesives. As a major wood component, native lignin is neither hygroscopic nor soluble in water. However, during technical pulping lignin becomes soluble in water, due to degradation and chemical changes.

The use of spent sulphite liquor (SSL) as an adhesive for paper, wood and other ligno-cellulosic materials is well-known in the art, and a large number of patent applications has been filed during the last three decades for the use of lignin products as adhesives for particle boards, plywood and fiber boards instead of conventional PF or UF adhesives. Reference is made to DE Patents Nos. 3 037 992, 3 621 218, 3 933 279, 4 020 969, 4 204 793 and 4 306 439 and PCT Applications published under Nos. WO 93/25622, WO 94/01488, WO 95/23232 and WO 96/03546.

The main drawback of using SSL as an adhesive for fiber board manufacture is its hygroscopicity. For this reason it cannot really compete with other natural or synthetic adhesives. When SSL is applied as an adhesive, it can be converted to an insoluble substance by curing. Chemically the curing of lignin is a cross-linking process, which leads to new carbon-carbon and ether bonds between different lignin molecules or within one macromolecule. Inter- as well as intramolecular cross-linking reactions decrease the solubility and swelling of lignin. Cross-links in lignin can be achieved either by condensation

or by radical coupling reactions. For the condensation reactions, either high temperatures and long heating times or mineral acids are required, which cause structural changes or charring in the wood particles. Recently, cross-linking of the lignosulphonate molecules by radical combinations has been developed. In most cases additional cross-linking agents for lignin are necessary, such as epoxides, polyisocyanates, polyols, poly(acryl amide)s, poly(ethylene imine) and aldehydes.

Further, it has been shown that laccase enzymes and other peroxidases can be used as polymerization or curing catalysts of lignin (DE Patent No. 3 037 992, WO 96/03546). However, the enzymes for creating radical reaction have shown limited success so far. Fibers and wood chips used in the production of fiber boards contain 5 - 20 % water and the laccases used need some water to effectively catalyze the polymerization reaction needed for extensive bonding of the fiberboard. Kraft lignin like native lignin to its major part is, however, insoluble in water and thus two solid phases are formed on the production line. An uneven distribution of the solids cause spotting and major failure in the strength properties of the board formed in the pressing stage.

A further problem relating to the use of isolated lignin is the high price of kraft lignin, which is near the limit for economical production of particle boards.

For the above mentioned reasons, lignin-based board production processes have not, so far, led to any major practical applications.

The present invention aims at eliminating the problems relating to the prior art. In particular it is an object of the present invention to provide a novel lignin-based adhesive for use with particle boards and similar wood-based products. It is another object of the present invention to provide new particle boards and similar wood-based products. It is a third object to provide a method for manufacturing the adhesive.

These and other objects, together with the advantages thereof over known lignin-based adhesives and processes for the preparation thereof, which shall become apparent from the

specification which follows, are accomplished by the invention as hereinafter described and claimed.

According to the present invention, lignin or lignin polymerized with oxidative enzymes or a soluble fraction of carbohydrates obtained from mechanical refining of lignocellulosic materials, is added to and mixed with the fibers or chips or flakes used as lignocellulosic raw material of the wood-based product in the form of a foam to provide an even dispersion of the solids.

Foamed UF and PF adhesives are known in the art. Said foams are used for improving process performance and product quality especially in adhesives with high solid contents. CA Patent Application No. 2,114,258 describes particle board production by using foamed mixtures of UF resins and animal blood. Also DE 3,644,067 describes the use of foamed materials in binding of fibers and flakes to produce a homogeneous adhesive application of fine particles (fibres or powders) on the furnish. T. Sellers describes in Forest Prod. J. 38 (1988) p. 55-56 the superior performance of foamed UF and PF resins in particle board manufacture especially at higher solids content of the adhesive. However, none of the prior art references mentions foaming of lignin-containing adhesive compositions. According to these references, the composition of the gas is not important either.

The lignin-based adhesive composition according to the present invention comprises an aqueous suspension of lignin and/or carbohydrates obtained from a process of pulping lignocellulosic materials. The suspension has been foamed to 1.05 to 10, preferably 1.1 to 8, in particular to 1.2 to 5 times the volume of the liquid mixture. It is preferred to use polymerized kraft lignin as the lignin component. Furthermore, according to the present invention, natural lignin in wood fibers is used to replace a substantial part of the lignin in the adhesive binder intended for particle board production. Thus, the present adhesive binders comprise an aqueous mixture of fibers together with lignin and/or carbohydrates obtained from a process of pulping lignocellulosic raw materials. The fibers in the adhesive binder as well as the added lignin and/or other lignocellulosic material radical polymerize due to the effect of the enzyme in an even oxygen-containing foam matrix. Instead of lignin,

the use of native lignin -containing fibers of MDF or TMP types leads to savings in adhesive use, and, however, to a corresponding strength.

The foam will homogenize the solid phase and the liquid phase, increase the viscosity of the mixture and prevent the sedimentation of the solids. The oxygen-containing foamed structure will also provide more oxygen for the oxygen-dependent oxidase catalyzed lignin/carbohydrate polymerization. Because of its large volume, the foam will cover more homogeneously the wood fibers and wood particles in the preparation of the boards. This will lead to better strength values and better control of the use of the adhesive in the production.

The invention will now be explained in more detail with the aid of the following detailed description and with reference to a number of working examples.

Within the context of the present invention, the terms “adhesive”, “adhesive binder” and “resin” designate a chemical composition which, in the wet stages of the manufacture of, e.g. particle and fiber boards, provides adhesion between the particles or fibers. After heat compression during board manufacture, the composition containing polymerized resin works as a binder which keeps the particles or fibers or flakes bonded together.

The term "wood-based product" denotes any lignocellulose-based product, such as particle boards, fiber boards (including high and medium density fiber boards, i.e. hard boards and MDF boards), flake boards, plywood and similar products constituted by particles, fibers or flakes of vegetable origin, in particular derived from wood or annular or perennial plants mixed with and bonded together with adhesive binders. According to a particularly preferred embodiment, the present invention is used for preparing particle boards and similar wood-based products in which wood particles can be crosslinked and joined together with the fibers of the adhesive composition, which will be attached to the particles by the lignin-based adhesive.

The term "fibers" encompasses organic and inorganic fibers of any suitable material having a length-to-thickness ratio of more than 6, preferably more than 10, in particular more than

20. It is preferred to use lignin-containing fibers, because the lignin of the fibers will be oxidized in the presence of oxidases and work as an adhesive in mixtures of wood particles and the adhesive foam. Particularly preferred lignocellulosic fibers are derived from wood or annular or perennial plants. Such fibers can be obtained from mechanical refining of wood or plants by refiner mechanical pulping, pressurized refiner mechanical pulping, thermomechanical pulping, groundwood or pressurized groundwood pulping, or chemi-thermomechanical pulping. Furthermore, fibers from the preparation of fiber boards (e.g. medium density or high density fiber boards) can be used in the preparation of particle boards.

In addition to the cellulosic fibers mentioned above, other natural fibers, such as cotton fibers, Abaca hemp fibers, sisal fibers, ramie fibers, flax fibers or jute fibers, can be used. Inorganic fibers such as glass fibers, carbon fibers, gypsum fibers, etc. can also be employed.

For polymerizing lignin and carbohydrates of soluble wood fractions, oxidative enzymes capable of catalyzing oxidation of phenolic groups can be used. These enzymes are oxidoreductases, such as peroxidases and oxidases. "Peroxidases" are enzymes which catalyze oxidative reaction using hydrogen peroxide as their substrate, whereas "oxidases" are enzymes which catalyze oxidative reactions using molecular oxygen as their substrate. Phenoloxidases (EC 1.10.3.2 benzenediol:oxygen oxidoreductase) catalyze the oxidation of o- and p-substituted phenolic hydroxyl and amino/amine groups in monomeric and polymeric aromatic compounds. The oxidative reaction leads to the formation of phenoxy radicals and finally to the polymerization of lignin and possibly the carbohydrate matter. In the method of the present invention, the enzyme used may be any of the enzymes catalyzing the biological radical formation and secondary chemical polymerization of low molecular weight lignins, such as laccase, tyrosinase, peroxidase or oxidase.

As specific examples of oxidases the following can be mentioned: laccases (EC 1.10.3.2), catechol oxidases (EC 1.10.3.1), tyrosinases (EC 1.14.18.1) and bilirubin oxidases (EC 1.3.3.5). Laccases are particularly preferred oxidases. They can be obtained from bacteria and fungi belonging to, e.g., the following strains: *Aspergillus*, *Neurospora*, *Podospora*,

Botrytis, *Lentinus*, *Polyporus*, *Rhizoctonia*, *Coprinus*, *Coriolus*, *Phlebia*, *Pleurotus*, *Fusarium* and *Trametes*.

Suitable peroxidases can be obtained from plants or fungi or bacteria. Preferred peroxidases are those which originate from plants, in particular horseradish peroxidase and soy bean peroxidase.

The terms "surfactant" or "surface active agent" are synonymously used to designate compounds which have affinity to water and to hydrophobic (e.g. fatty) materials, thus aiding the hydrophobic materials to suspend in water.

Adhesive component

As mentioned above, lignin of different origins can be used as the adhesive component of the present foamed adhesive binders. In particular, isolated lignin of, e.g., the sulphate, sulphite, ORGANOSOLV and MILOX processes can be used.

However, in addition to lignin or instead of it, various soluble wood fractions can also be employed. Thus, it is known in the art that during mechanical refining of chips, a part of the compounds of the fibrous raw material is dissolved (about 1 % of the fiber weight). This fraction contains primarily the same chemical components as the fibers (carbohydrates, extractives and lignin). To mention an example: the soluble fraction of softwood chips contains some 40 to 70 % carbohydrates, 10 to 25 % lignin and 1 to 10 % extractives, whereas hardwood pulping process water contains some 20 to 60 % carbohydrates, 10 to 25 % lignin and 10 to 40 % extractives.

It has been found that this soluble lignin/carbohydrate fraction obtainable from mechanical or chemimechanical pulping is particularly useful as an additive or adjuvant for gluing of particles boards, fiber boards and other similar wood-based composite products. Particularly good gluing is achieved if this fraction (as is the case with lignin) is polymerized with laccase (or similar) oxidase enzyme(s). The results are on the same level as those obtainable with conventional phenol or urea formaldehyde resins.

It should be noticed that a similar soluble fraction can be obtained by treating lignocellulosic raw material, such as wood fibers or saw dust, with hydrolases, e.g. cellulases, hemicellulases and pectinases.

Manufacture of adhesive binder foam

The present invention relates to foam-based technology for providing good dispersion of the different solids. Thus, efficient contact during economical production of particle board is achieved using adhesive binders based on oxidase-activated lignin and/or carbohydrates together with wood fibers or other lignocellulosic fibers. 0.1 to 50 %, preferably 0.5 to 30 % and in particular about 1 to 10 % by weight of the kraft lignin or similar lignin is replaced by fibers in the foam-based, oxidase-activated adhesive formulation.

According to the preferred embodiment of the present invention, in which wood-derived fibers are used, lignin and the fibers are activated by oxidases are mixed with the chips in the form of a foam that provides a good viscose suspending medium to form an even dispersion of the solids and hence improves substantially the quality of the particle board formed. The use of the foamed adhesives as described in this invention leads also to better control of the application and to substantial savings in adhesive use. The foam is produced by the use of a surface active agent.

The foamed, activated fiber and lignin/carbohydrate dispersion can be produced separately from the chips, which are mixed into the foam by known mixing technology, e.g. by extruding or spraying the foam onto the fibers or chips. The foam can also be produced simultaneously by mixing the fibers and chips with the foam chemicals, the lignin/carbohydrates and the laccase.

According to the present invention, the foam is produced by dispersing the mixture of lignin and fibers into water to form a suspension and bubbling a gas through the suspension to form bubbles having a medium diameter of 0.001 to 1 mm, in particular about 0.01 to 0.1 mm.

The foam is produced by the use of a surface active agent that can be anionic, cationic or non-ionic. Thus, the surfactant can be selected from the group consisting of alkylsulfonate or alkyl benzene sulfonate, Tween® and other commercial compounds, fatty acid soaps, lignosulfonates, sarcosinates, fatty acid amines or amines or poly(oxyetylene alcohol)s and wood and plant extractives. Foam stabilizers and solid surfactants, such as CMC, gelatin, pectin, wood extractive and similar compounds, can be used to produce and enhance the foam stability. A small amount of the surface active compounds is needed, i.e. about 0.01 to 10 %, in particular about 0.05 to 5 %.

The foam can be produced by foaming in a static foamer or in a turbulent foam cell by using known mixing technology. Although any gas can be used for foaming, it is preferred to use oxygen-containing gases, such as air, oxygen enriched air, oxygen or pressurized systems of these. The importance of using oxygen-containing gases is discussed in further detail below.

The foam produced is essentially stable during handling, storage, transport and manufacture of wood-based products and it has a density in the range of 0.1 to 0.9 kg/l, in particular about 0.2 to 0.7 kg/l, and a medium foam bubble diameter in the range of 0.005 to 0.1 mm, in particular about 0.01 to 0.1 mm, preferably about 0.02 mm.

The method according to the present invention can be used for all oxidase-catalyzed, previously unsuccessfully suggested enzyme-catalyzed glueing processes using oxidases. The enzyme used can be any of the enzymes catalyzing the oxidation and polymerization of aromatic compounds or lignins, such as laccase, tyrosinase, or other oxidases, as mentioned above. The amount of enzyme used varies depending on the activity of the enzyme and on the amount of dry matter content of the composition. Generally, the oxidases are used in amounts of 0.001 to 10 mg protein/g of dry matter, preferably about 0,1 to 5 mg protein/g of dry matter. The activity of the oxidase is about 1 to 100,000 nkat/g, preferably over 100 nkat/g.

In connection with the present invention it has been found that oxygen plays a decisive role in the enzymatic polymerization of lignin of any origin. This is important in particular for the production of adhesives for the manufacture of fiber boards, particle boards and flake

boards and other similar wood-based products. Thus, in addition to the aromatic reactant, also oxygen is needed. The oxidative reaction leads to the formation of phenoxy radicals and finally to the polymerization of lignin.

In the known methods discussed above, crosslinking was only partially achieved because of apparent limitations on the availability of oxygen. The limitation of the reaction by oxygen manifests itself in the long reaction times used, and in the poor strength properties obtained, thus impairing the result of the enzyme-aided polymerization.

Oxygen can be supplied by various means, such as efficient mixing, air enriched with oxygen or introducing oxygen supplied by enzymatic or chemical means to the solution. However, according to the present invention, by foaming the suspension with an oxygen-containing gas it is possible to provide the oxygen needed for full polymerization of the lignin. At the same time, the foamed structure gives enough strength to the adhesive so as to prevent sedimentation of water-insoluble lignin or the polymerized lignin from the adhesive thus producing failure in the production and in the board product.

The present invention provides considerable advantages. Thus, the foamed structure can be utilized in the preparation of particle boards of lignin and/or carbohydrates or polymerized lignin by utilizing the adhesive properties of the lignin or polymerized lignin, whereas the fibers work as crosslinkers between the particles. The foam will homogenize the inhomogenous solid phase and the liquid phase, increase the viscosity of the mixture and prevent the sedimentation of the solids and the fibers. The oxygen-containing foamed structure will also provide more oxygen for oxygen-enhanced oxidase catalyzed lignin/carbohydrate polymerization. Because of its large volume the foam will cover more homogeneously the wood fibres and wood particles in the preparation of the boards. The adhesive binder is evenly distributed throughout the wood-based product, about $0.02 \pm 0.002 \ldots 0.2 \pm 0.02$ g/cm^3, preferably 0.04 to 0.08 g/cm^3 in any randomly selected volume unit of the product.

This leads to better strength values and better control of the use of the adhesive in the production.

The following non-limiting working examples illustrate the invention.

Example 1
Manufacture of MDF fibers

MDF fibers were prepared from hardwood (Swedish birch) by refining birch chips in a conventional TMP process at a refiner pressure of 4 bar.

The fiber quality was tested using both Bauer McNett analysis and a Pulmac shive analysis. The results are shown below:

Table 1. Fiber characteristics

Bauer McNett	
> 16 (>1.19 mm)	18.6
16-30 (1.19-0.595 mm)	25.1
30-50 (0.595-0.297 mm)	12.4
50-100 (0.149 mm)	29.0
<100 (<0.149 mm)	15.0
Pulmac shive analysis	
Screen plate 0.15 mm	7.0
Screen plate 0.25 mm	0.8

Example 2
Manufacture of particles boards

This Example describes the preparation of a foamed adhesive consisting of kraft lignin, laccase, wood fibre and the manufacture of particle boards from the adhesive. The test has been carried out on laboratory scale.

Kraft lignin Indulin AT and 1 - 10 % of its weight substituted with the MDF fibers of Example 1 were used as a binder mixture in particle board test panel manufacture.

3.6 - 3.96 g of the lignin was vigorously mixed with 0.04 - 0.4 g of MDF fibers and aerated for 30 min with 4.0 g of laccase concentrate (activity 4,000 nkat/g) in 2.0 g of 2 M sodium acetate buffer (pH 4.5). 1.4 g of the mixture was mechanically mixed with 4.4 g of chips. The chips had already been treated with 1 % of wax (Mobilex 54, 60 % emulsion) of the dry weight of the fibers. The reference tests were performed without laccase (water was used instead) and using commercial ureaformaldehyde resins.

For strength tests particle board panels of the size of 50 mm x 50 mm x 2 mm (weight about 5 g) were prepared by pressing 2 min in 30 kp/cm^2 of pressure and 190 °C of temperature. After pressing, the panels were then cut into four pieces (50 mm x 12 mm x 2 mm). These pieces were tested for parallel tensile strength with Zwick tensile strength testing equipment.

Table 2. Results of glueing tests for particle boards

Foamed binder	**Tensile strength MPa**
Water	3.0 ± 0.3
Indulin AT (10 %) + MDF fibers (90 %)	8.5 ± 0.4
Indulin AT (10 %) + MDF fibers (90 %) + laccase	11.9 ± 0.7
Indulin AT	7.2 ± 0.4
Indulin AT + laccase	12.7 ± 0.6
Reference UF resin	12.5 ± 0.7

As apparent from the results of Table 2, up to 10 % of laccase-activated kraft lignin can be substituted as an adhesive by MDF fibers without any significant loss in tensile strength. The strength properties of particle boards prepared according to the present invention are quite comparable to those of conventional UF resin glued boards.

WHAT IS CLAIMED IS:

1. A lignin and/or carbohydrate -based adhesive binder composition for particle boards and similar wood-based products, wherein
- the composition comprises a mixture of lignin-containing fibers and lignin and/or carbohydrates obtained from a process of pulping lignocellulosic materials, and oxidative enzymes capable of catalyzing oxidation of phenolic groups, and
- the mixture is being foamed to 1.1 to 10 times the volume of the liquid mixture.

2. The composition according to claim 1, wherein the lignin comprises polymerized kraft lignin.

3. The composition according to claim 1, wherein the foam has a solids content of 1 - 90, preferably about 5 - 70 % by weight.

4. The composition according to claim 1, wherein the density of the foam is in the range from 0.1 to 0.9 kg/l, in particular about 0.2 to 0.7 kg/l, and the medium foam bubble diameter is in the range of 0.0001 to 0.1 mm, in particular about 0.01 to 0.05 mm.

5. The composition according to any one of the claims 1 to 4, wherein the fibers are lignocellulosic fibers derived from wood or annual or perennial plants.

6. The composition according to claim 5, wherein the fibers are obtained from mechanical refining of wood or plants by refiner mechanical pulping, pressurized refiner mechanical pulping, thermomechanical pulping, groundwood or pressurized groundwood pulping, or chemithermomechanical pulping.

7. The composition according to claim 6, wherein the fibers are obtained from the preparation of fibers for fiber boards.

8. The composition according to claim 1, wherein the fiber content is 0.1 to 50, preferably 0.5 to 30 and in particular 1 to 10 % of the weight of the lignin.

9. The composition according to claim 1, wherein the foam contains surfactants in an amount of 0.1 to 10, preferably 0.2 to 5 % by weight of the aqueous suspension.

10. The composition according to any one of the preceding claims, wherein the composition contains an oxidase or a peroxidase, the amount of the oxidase being 0.1 to 5 mg protein/g dry matter.

11. A method of preparing a lignin and/or carbohydrate -based adhesive binder for particle boards and similar wood-based products, comprising the steps of

– using lignin and/or carbohydrates obtained from a pulping process of lignocellulosic raw materials as raw material,
– using fibers based on wood or annual or perennial plants as raw material,
– forming an aqueous suspension of the lignin and/or carbohydrates and the fibers, and
– foaming the aqueous suspension to provide a foamed adhesive binder composition.

12. The method according to claim 11, comprising the steps of

– forming an aqueous suspension containing 1 - 90 % by weight of solid matter comprising lignin, fibers and surfactants, and
– foaming the aqueous suspension with an oxygen-containing gas to 1.1 - 10 times the volume of the suspension to provide a foam having a solids content of 5 - 70 % by weight.

13. The method according to claim 11 or 12, wherein the lignin comprises polymerized kraft lignin.

14. The method according to claim 12, which comprises the step of forming an aqueous suspension containing lignin, fibers, an oxidase and surfactants.

15. Particle board or a similar wood-based product comprising wood particles, fibers or flakes mixed with an adhesive binder according to any one of the claims 1 to 10.

16. A method of manufacturing a particle board or a similar wood-based product, comprising the step of mixing wood particles, wood fibers or wood flakes with a lignin and/or carbohydrate based adhesive binder according to any one of the claims 1 to 10 so as to incorporate 2 - 20 % of the solids of the adhesive binder composition into the wood-based product.

INTERNATIONAL SEARCH REPORT

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A. CLASSIFICATION OF SUBJECT MATTER

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B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

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Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

SE,DK,FI,NO classes as above

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

WPI, CAPLUS

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 9325622 A1 (NOVO NORDISK A/S), 23 December 1993 (23.12.93), page 1, line 15 - page 4, line 2, example 3; claims --	1-16
A	WO 9220857 A1 (CALL, HANS-PETER), 26 November 1992 (26.11.92), abstract, page 4, last paragraph - page 10, second paragraph; example 4; claims --	1
A	EP 0492016 A1 (K.C. SHEN TECHNOLOGY INTERNATIONAL LTD.), 1 July 1992 (01.07.92), page 3, line 37 - line 45, abstract, claims --	1-16

[X] Further documents are listed in the continuation of Box C. [X] See patent family annex.

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International application No.
PCT/FI 98/00026

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